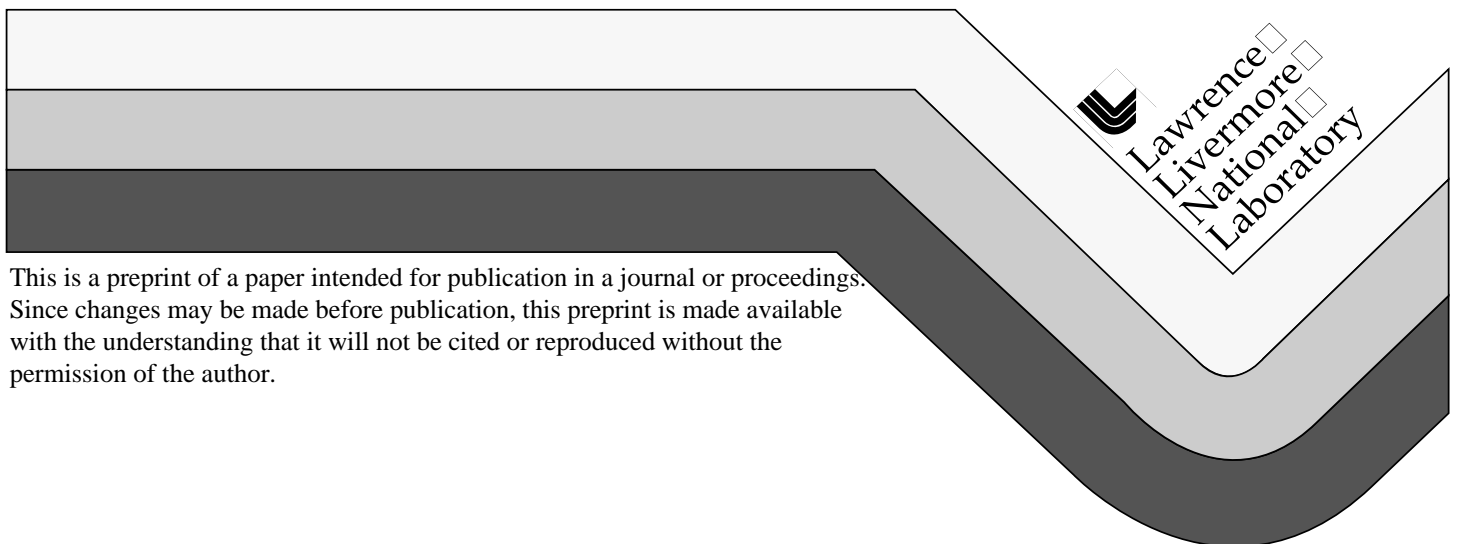


**Radionuclide Characterization and Associated Dose from
Long-Lived Radionuclides in Close-In Fallout Delivered to
the Marine Environment at Bikini and Enewetak Atolls**

William L. Robison
Victor E. Noshkin

This paper was prepared for submittal to
The Science of the Total Environment
Conference Proceedings for *International Symposium*
on Marine Pollution, 5–9 October,
Monte-Carlo, Principality of Monaco

September 1998



DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Radionuclide Characterization and Associated Dose from Long-Lived Radionuclides in Close-In Fallout Delivered to the Marine Environment at Bikini and Enewetak Atolls

William L. Robison¹, and Victor E. Noshkin²,

¹Lawrence Livermore National Laboratory, P.O. Box 808, L-286
Livermore, CA 94551-0808
Phone: (925) 422-3885
FAX: (925) 423-6785

²Lawrence Livermore National Laboratory, P.O. Box 808, L-396
Livermore, CA 94551-0808
Phone: (925) 422-0911
FAX: (925) 424-3255

Abstract

Between June 1946 and October 1958, Enewetak and Bikini Atolls were used by the United States as testing grounds for 66 nuclear devices. The combined explosive yield from these tests was 107 Mt (Mt TNT equivalents). This testing produced close-in fallout debris that was contaminated with quantities of radioactive fission and particle activated products, and unspent radioactive nuclear fuel that entered the aquatic environment of the atolls. Today, the sediments in the lagoons are reservoirs for 10's of TBq of the transuranics and some long-lived fission and activation products. The larger amounts of contamination are associated with fine and coarse sediment material adjacent to the locations of the high yield explosions. Radionuclides are also distributed vertically in the sediment column to various depths in all regions of the lagoons. Concentrations greater than fallout background levels are found in filtered water sampled over several decades from all locations and depths in the lagoons. This is a direct indication that the radionuclides are continuously mobilized to solution from the solid phases. Of particular importance is the fact that the long-lived radionuclides are accumulated to different levels by indigenous aquatic plants and organisms that are used as food by resident people. One might anticipate finding continuous high contamination levels in many of the edible marine organisms from the lagoons, since the radionuclides associated with the sediments are not contained and are available to the different organisms in a relatively shallow water environment. This is not the case. We estimate that the radiological dose from consumption of the edible parts of marine foods at Enewetak and Bikini is presently about 0.05% of the total 50-year integral effective dose from all other exposure pathways that include ingestion of terrestrial foods and drinking water, external exposure and inhalation. The total radiological dose from the marine pathway is dominated by the natural radionuclides, ²¹⁰Po and ²¹⁰Pb. Man-made radionuclides presently contribute less than 0.3% of the dose from these natural radionuclides in the marine food chain and within about 90 y only 0.05%.

Key Words: Marshall Islands; Radioactivity; Dose assessment; Marine environment

Introduction

Bikini and Enewetak are 2 of the 29 atolls and 5 islands that make up the Republic of the Marshall Islands located in the equatorial Pacific roughly 2,500 nautical miles southwest of the Hawaiian Islands. Bikini Atoll is located at 11°35'N 165°23'E and consisted of 26 (23 remaining) coral islands (8.8 km² in total area, omitting intertidal areas) located on a reef 78 km² in area surrounding a lagoon 630 km² in area. The mean depth of the lagoon is 46 m. Enewetak's center is at 11°30'N 162°15'E. The Atoll originally consisted of a ring of 42 (39 remaining) low islands 6.9 km² in area arranged on a roughly elliptical shaped reef 83 km² in area. The lagoon has an area of 932 km² and a mean depth of 48 m (Emery et al. 1954). The United States conducted 2 nuclear tests in and above Bikini lagoon in 1946. The following year the U.S. Atomic Energy Commission took steps to establish proving grounds in the Pacific for testing atomic weapons. Enewetak Atoll was selected and designated as the Enewetak Proving Grounds (EPG) where testing of nuclear devices began in 1948. In 1953, the status of Bikini Atoll was also changed to that of a proving ground. The two atolls were then to be known as the western part of the United States Pacific Proving Grounds (PPG). During 7 of the years between 1946 and 1958, the United States exploded 24 and 42 nuclear devices either on the surface or underwater in the lagoon, or on the surface or above land at Bikini and Enewetak Atolls, respectively. Two additional nuclear tests were conducted in and over the ocean outside, but in the vicinity of the atolls. Prior to any testing at either atoll, the original residents were relocated to other islands in the Marshall Islands.

The total combined explosive yield at the atolls was 107 Mt (Mt TNT equivalent) or approximately 20% of the estimated total yield from all atmospheric nuclear testing. Each successful nuclear test produced mixtures of over 200 radioactive fission products representing elements from nickel to ytterbium (Nethaway and Barton, 1973); a variety of short and long-lived particle-activated products; and quantities of unspent nuclear fuel. These radionuclides became associated with materials incorporated into the fireball of the explosions. The characteristics of the particles differed with each test and were determined by the yield and the types of material associated both with the device and in the environment of the explosion. Adams et al. (1960) discuss in some detail the composition and different properties of the radioactive fallout particles that were generated from tower, ground surface, and over water nuclear detonations.

A small fraction of the condensed material, and associated radioactivity, resulting from each test was deposited to the terrestrial and marine environments of Bikini and Enewetak Atolls. The radionuclides deposited on the surface

of the lagoon either settled rapidly to the bottom sediments in association with particles, or remained as dissolved or particulate-associated species in the water column, and were eventually discharged to the equatorial Pacific Ocean.

The U.S. testing moratorium began on 31 October 1958 and marked the end of testing at the western PPG. The contaminated sediments in the lagoon are now a source for a variety of different radionuclides (Nelson and Noshkin, 1973; Nevissi and Schell, 1975; Noshkin, 1980; Schell, et al. 1980; Noshkin et al. 1997a; 1997b; 1998a; 1998b). Specific radionuclides are continuously remobilized from old and fresh carbonate sediments to the interstitial and overlying seawater. A number of physical, chemical and biological processes are responsible for this diagenesis. All radionuclides are accumulated to different degrees in tissues and organs of benthic, pelagic, and reef dwelling organisms. Radioactive decay, and loss resulting from natural processes, has reduced the inventories of all but the longer-lived radionuclides. Today the exposed sediments at Bikini and Enewetak are still reservoirs for 10's of TBq of $^{239+240}\text{Pu}$ with lesser amounts of ^{241}Am , ^{238}Pu and some other long-lived fission and activation products such as ^{90}Sr , ^{137}Cs and ^{207}Bi to name a few.

The radionuclides associated with the components in the lagoon sediments are not contained and are available to different aquatic organisms in a relatively shallow water environment. One might then anticipate finding continuous high contamination levels in many of the edible marine organisms from the lagoons. This is not the case. In this report we discuss some of the environmental characteristics of several longer lived radionuclides still found in the near shore, and deep lagoon environments that can be accumulated by reef and pelagic organisms used as food thus resulting in a radiological dose to the local inhabitants. It is the presence of the longer-lived man-made isotopes in the environment that are of a concern to the people resettling Enewetak and Bikini Atoll.

Nuclear Testing at Bikini and Enewetak Atolls

The total yield for all tests conducted at the PPG was 107 Mt with an estimated fission yield of approximately 57.1 Mt (Gallery of U.S. Nuclear Tests, 1997). At Bikini the total yield was 76.8 Mt and approximately 50% of this value was fission yield. At Enewetak the total yield was only 30.2 Mt, however, approximately 63% of this value was from fission. Two tests at Enewetak resulted in no nuclear yield, but the high explosives scattered fissionable material over the surrounding areas. Five tests at Bikini were classified as fissions (Gallery of U.S. Nuclear Tests, 1997) where yields were far below those predicted. Four of the larger tests conducted during Operation Castle in 1954 at Bikini resulted in yields significantly higher than predicted (Gallery of U.S. Nuclear Tests, 1997).

Narratives about the different test series, purposes, yields, description of the devices, and other information can be found in Hawthorne, 1979; Hansen, 1988; Gallery of U.S. Nuclear Tests, 1997; and Simon and Robison, 1997.

Radionuclides Detected in the Marine Environment

During the early years of testing, a variety of different marine samples such as plankton, fish, invertebrates, seawater, and sediments were collected from the lagoons to assess the relative levels of gross radioactivity associated with the samples (Welander, 1957; Bonham, 1959). Chemical separations were conducted to identify specific radionuclides associated with the marine samples following the Castle series in 1954. Some fission products and the activated radionuclides ^{54}Mn , ^{55}Fe , ^{59}Fe , ^{57}Co , ^{58}Co , ^{60}Co , ^{65}Zn , ^{185}W , ^{239}Np and ^{237}U (Lowman, et al., 1957; Weiss and Shipman, 1957; Lowman, 1960) were identified and measured in plankton, parts of fish, and invertebrates collected from locations within and outside Bikini and Enewetak lagoons.

The first major aquatic survey that developed quantitative data for different radionuclides in marine samples from both atolls was conducted during 1964, 6 y after the moratorium on testing at the PPG (Welander, et al., 1967; Welander, 1969). Gamma spectroscopy using sodium iodide detectors and spectrum stripping methods were used to determine the concentrations of ^{54}Mn , ^{57}Co , ^{60}Co , ^{65}Zn , ^{106}Ru , ^{125}Sb , ^{137}Cs , and ^{207}Bi in 3 bottom sediment samples, 5 samples of seawater, plankton, and parts of fish and invertebrates. Radiochemical separations provided information on concentrations of ^{55}Fe , ^{90}Sr , $^{102\text{m}}\text{Rh}$, ^{144}Ce , ^{155}Eu , $^{110\text{m}}\text{Ag}$ and $^{239+240}\text{Pu}$, and Koranda (1965) determined levels of ^{14}C and ^3H in different marine samples. By the early 1970's, the radionuclides with relatively short half-lives such as ^{54}Mn , ^{57}Co , ^{65}Zn , ^{144}Ce , $^{110\text{m}}\text{Ag}$, ^{95}Zr , and ^{106}Ru had sufficiently decayed so that they were only occasionally detected in sediment or biota from the lagoons. There were several additional long-lived isotopes identified in marine samples collected on subsequent sampling expeditions to the atolls. During these latter programs, Ge (Li) detection systems were used, and it was possible to resolve the concentrations of other gamma emitting radionuclides present in the samples. Radiochemical separations with low-background detection systems made possible the identification of other long-lived, non-gamma-emitting radionuclides such as ^{237}Np , ^{241}Pu , ^{244}Cm , ^{63}Ni , ^{99}Tc , ^{151}Sm , and ^{147}Pm to name a few. An extensive radiological survey of the marine environment at Enewetak was conducted in 1972-73 in preparation for resettlement of the atoll. Fish, invertebrates, plankton, sediment and seawater were collected from the lagoon and analyzed for many different radionuclides (Nelson and Noshkin, 1973). Robison (1973) used these data along with diet information, and concluded that the majority of dose from ingestion of edible parts of marine food at the time was from radionuclides such as ^{137}Cs , ^{60}Co , and ^{90}Sr .

Radionuclides Associated with Lagoon Sediments

The radionuclides have not remained fixed to the original fine material deposited to the sediments during testing. They are found associated with all major coarse components (such as Foraminifera, coral, remains of Halimeda algae, and shells of mollusks) in the surface and subsurface sediments. Release of the radionuclides results from the action of a number of different biological-chemical-physical transformations, making possible their diffusion along chemical potential gradients in the interstitial solution leading to penetration further into the sediment column, or release to the lagoon seawater at the sediment-water interface. Some fraction of the released radionuclides are capable of being adsorbed, or absorbed by other larger living or dead components in the sediment column.

Major sediment collection programs were conducted at the atolls in 1972 and 1979. In the latter program, a more systematic sampling grid was established at both atolls to provide a series of dimensionally comparative surface sediment samples from all regions of both lagoons. Concentration and inventories for all detectable gamma-emitting radionuclides were determined in the fine (<0.5 mm) and coarse (>0.5mm) sediment components (Noshkin et al. 1997a; 1997b; 1998b). During this time, the dominant gamma-emitting radionuclides measured in the sediment samples were ^{241}Am , ^{207}Bi , ^{60}Co , ^{155}Eu , and ^{137}Cs . Radionuclides such as $^{152,154}\text{Eu}$, ^{125}Sb , and $^{102\text{m}}\text{Rh}$ were occasionally detected in samples from the more contaminated regions of the lagoon. Mean concentrations of all radionuclides in the 0–2 and 0–4 cm section of surface sediment were nearly identical indicating the radionuclides were well mixed in the sediment of a depth of at least 4 cm.

Contours for ^{241}Am concentration associated with the fine and coarse fractions in Bikini surface (0-2 cm) sediment are shown in Figure 1. The contours for ^{241}Am and ^{207}Bi concentrations in the fine surface sediment at Enewetak are shown in Figure 2. At Bikini it is evident that ^{241}Am with fine and coarse sediment is distributed non-uniformly over the surface of the lagoon floor. Concentrations range from 10 to over 12000 Bq kg⁻¹ with 1100 and 600 Bq kg⁻¹ being the average concentration associated with the fine and coarse components, respectively. The highest concentrations are associated with surface deposits from the northwestern quadrant of the lagoon, adjacent to the locations of the larger explosions (see Fig. 1), and the concentration for both the fine and coarse fractions decrease from the north to the south, and from the west to the east.

The distribution of ^{241}Am with the fine surface sediment in Enewetak is not as widespread as at Bikini, as shown in Figure 2. The highest concentrations are associated with surface sediments on the western side of the atoll

in the region lagoonward of the Oak, Mike-Koa craters. A second region of relatively high concentration is found with the fine surface sediments, lagoonward of Runit Island (E-24) on the east reef. At Enewetak, the surface concentrations range from <1 to 1300 Bq kg^{-1} with 170, and 130 Bq kg^{-1} being the average concentration associated with the fine and coarse fractions, respectively.

The difference in average concentration in the surface sediment between Bikini and Enewetak is a reflection of the difference in the yields at the atolls. Distributions for the concentrations of other major radionuclides in fine and coarse sediment resemble the gross features of ^{241}Am but have some very specific differences. For example, the concentration of ^{207}Bi associated with the fine fraction of surface sediment decreases in a north-south direction much like ^{241}Am as shown in Figure 2. However, note that the highest levels of activity are associated with the sediments south of Boken-Enjebi (E 9-10) islands, rather than the sediments from the general area lagoonward of Oak, Mike-Koa test sites as found for ^{241}Am . Differences are also found comparing the distributions for $^{239+240}\text{Pu}$ with ^{238}Pu at Enewetak (Noshkin et al. 1998a), and for example, ^{137}Cs with ^{155}Eu , ^{60}Co , ^{90}Sr , or ^{241}Am at both atolls (Nelson and Noshkin 1973; Noshkin, 1980; Schell et al. 1980; Noshkin et al., 1997a; 1997b; 1998b). After 40 years the radionuclides have not been well mixed throughout the lagoon surface deposits.

Inventories of the radionuclides in the total surface sample (fine + coarse fraction) were determined from contour maps generated by kriging (a specific gridding method used to develop surface contour plots) associated with the Surfer® (Surfer for Windows, Golden Software, Inc., 809 14th St., Golden, CO 80401-1866) software. All previous reported inventories (determined from the 1972 and 1979 data) at Enewetak and Bikini (Nelson and Noshkin, 1973; Noshkin 1980; Noshkin et al., 1997a; 1997b) were recomputed using kriging techniques, and normalized to a depth of 2 cm for relative comparison. Results are shown in Table 1. The inventory for $^{239+240}\text{Pu}$ and ^{90}Sr at Enewetak was only determined in 1972 and we do not have a sufficient database to estimate the total inventory of ^{90}Sr at Bikini. Radionuclide inventories are shown for the date of collection and then decay corrected to October 1998. Total inventory values are divided by the area of each atoll to provide the average GBq m^{-2} associated with the surface 2-cm section of sediment.

The inventories associated with the surface sediments are considered very reliable. However, estimates of the amounts distributed to depth in the lagoon are not as certain. Over the years, 44 sediment cores of different length have been collected in the lagoons outside the craters, and away from test site locations within the atolls. The hard carbonate substrate made core sampling very difficult. The mean depth of the 44 cores was $18 \pm 6 \text{ cm}$. The

radionuclides appeared to be well mixed to a depth of about 9 cm. Below about 10 cm the radionuclide concentrations decrease with depth, and consequently, the inventories increase slowly with depth. The inventory of all radionuclides in the 0–2 cm layer compared to the total (to depths of 10–30 cm) in the cores ranged from 2 to 25% and averaged 13 ± 5 . This percentage was applied to the entire lagoon to estimate the total inventory and activity per unit area to 30 cm shown in Table 1. The amounts represent minimum values because in some areas of the lagoon the radionuclides may be mixed to deeper depths (McMurty et al., 1985).

In the surface sediments, the total and average inventory of the transuranics ($^{239+240}\text{Pu}$ and ^{241}Am) and the fission product, ^{155}Eu , at Bikini exceed the amount at Enewetak. The amounts of ^{137}Cs and ^{60}Co in the sediments of both atolls are not substantially different while there is approximately 10 times more ^{207}Bi at Enewetak. The water column inventory of ^{90}Sr at Bikini is approximately 1.6 times the amount found in Enewetak (Noshkin et al., 1974). Therefore, the amount of ^{90}Sr is expected to be proportionally higher in the sediments from Bikini Atoll. The mean inventories for ^{137}Cs , ^{155}Eu , and ^{60}Co in the surface sediments decreased between 1972 and 1979. Comparing the decay corrected results it would appear that any reduction of the transuranics in the sediments is principally through radioactive decay. Only small quantities of these radionuclides would be mobilized from the sedimentary reservoir to the water column over time. Decay and natural processes leading to mobilization and migration act to reduce the amount of ^{137}Cs , especially at Enewetak, and ^{60}Co at both atolls.

Radionuclides in Lagoon Seawater

Lagoon water at both atolls exchanges with the surface waters of the North Equatorial Pacific Ocean through the major channels and passages in the reef. The estimated mean residence time of the seawater in the lagoons is from 1 to several months (Von Arx, 1948; Atkinson et al. 1981). Therefore, following the last nuclear test at Enewetak in 1958, the residual radionuclides deposited over the lagoon either settled to the bottom, or remained as dissolved or particulate species in the water, and were within months discharged to the ocean. Accepting the thesis that the radionuclides associated with the sediment remain immobile after deposition, then any activity in the water column subsequent to 1958–59 should only equal the fallout levels in the north equatorial Pacific surface water. However, wherever and whenever over several decades water was sampled in the lagoons or on the reef, the concentrations of $^{239+240}\text{Pu}$ and ^{137}Cs , for example, greatly exceeded the fallout background levels in the Equatorial Pacific surface waters. Table 2 summarizes arithmetic mean concentrations, and the range in concentrations for $^{239+240}\text{Pu}$ and ^{137}Cs in seawater samples collected during different periods from the regions of Enewetak and Bikini.

Also shown for comparison are concentrations in surface water sampled in the North Equatorial Pacific near the atolls, and well away from the influence of the atolls. These results are a direct indication that plutonium and ^{137}Cs , and other radionuclides such as ^{241}Am , ^{90}Sr , ^{60}Co , ^{155}Eu , and ^{207}Bi (Nelson and Noshkin, 1973), are continuously released from the solid phases to solution in the atolls, and are redistributed by currents to other regions within and outside the atolls as soluble and particulate species. Both the reduced and the oxidized forms of $^{239+240}\text{Pu}$ coexist in the lagoon water, with oxidized forms generally dominating. On the average, 78% of the total plutonium in solution in the lagoons and over craters was in the oxidized state. Over 90% of the $^{239+240}\text{Pu}$ found associated with sedimentary or particulate material in the lagoon were in the reduced state. Therefore, after desorption of reduced and oxidized species, the reduced state disappear from the solution either by oxidation to the higher state or loss by adsorption onto newly exposed surfaces. Since the reduced forms of plutonium are ubiquitous in nearly all samples, it is possible that some of the reduced forms are stabilized by complexation to some degree, but that the stability of the complex may change with time.

Between 1972 and 1982 the average “soluble” $^{239+240}\text{Pu}$ in Enewetak lagoon, determined from the 131 samples in Table 2, was 0.78 Bq m^{-3} . At Bikini the mean determined for the 71 lagoon samples over this period was 1.54 Bq m^{-3} . Taking into account the dimensions for each lagoon, these mean concentrations convert to standing inventories of 35 and 44 GBq in the lagoon water mass at Enewetak and Bikini, respectively. These quantities, which exclude the $^{239+240}\text{Pu}$ associated with suspended particles, represent 0.04-0.06%, respectively, of the sediment inventories to a depth of 30 cm shown in Table 1.

The mean concentrations of ^{137}Cs at Enewetak and Bikini in Table 2 are corrected for global fallout background (5 Bq m^{-3}) and decayed to 10/98. A mean concentration of $6 \pm 2 \text{ Bq m}^{-3}$ is determined at Bikini from the 11/72, 1/77 and 9/82 data. At Enewetak the decay corrected mean concentration for 1972, 1974, 1975, and 1976 is $4 \pm 1 \text{ Bq m}^{-3}$. These convert to lagoon inventories during 10/98 of approximately 170 GBq, and represent from 4 to 5 % of the sediment inventory estimated to 30cm. ^{137}Cs , as expected, is released to solution and lost from the atoll environment at a rate much more rapid than plutonium. The mean level ^{137}Cs in lagoon seawater is presently only twice the value of “global fallout” levels found in California Pacific coastal surface waters (Wong et al., 1992) in the late 1980’s.

Concentrations in Edible Marine Organisms

There are more than 700 species of fish in the Marshall Islands lagoons, but only a few have been routinely collected over the years for radionuclide analysis. Several species of reef fish are abundant in all regions of the lagoons, easy to catch, territorial and preferred in the Marshallese diet. These include surgeonfish (Acanthurus triostegus), a grazing herbivore; and mullet (Crenimugil crenilabis and Neomyxus chaptalii), herbivorous detrital feeders, and goatfish (Mulloidichthys samoensis), that consume benthic fauna, including small clams, crustaceans, and small fish. Other reef fish occasionally collected included parrotfish, triggerfish, and flagtail.

All reef fish were collected using throw nets at accessible locations on the lagoon and ocean side reef of the islands. The larger predator pelagic (bonito, mackerel, tuna, jacks), and benthic species (groupers, snappers) that roam the lagoons were also collected using sport fishing gear while trolling in the lagoons. These fish make up part of the marine food diet. Selected invertebrates that are sometimes eaten were also collected for analysis. All samples were dissected to allow analysis of the radionuclides associated with different tissues and organs so that the concentration of the radionuclides in any combination of tissues and organs could be reconstructed if necessary. However, special attention was given to the muscle tissue since it was determined that flesh was usually the only tissue of fish consumed by residents of the Marshall Islands. It was particularly important to provide flesh-only samples for analysis because orders of magnitude difference exist between the flesh, and whole body concentrations of radionuclides such as ^{90}Sr , $^{239+240}\text{Pu}$, and ^{241}Am that are most concentrated in nonedible parts of the fish. Whereas the difference between the flesh, and whole body concentration of ^{137}Cs is insignificant, less than 1 and less than 3% of the transuranics (plutonium and americium) and ^{90}Sr body burdens, respectively, are found in the flesh of any fish. For ^{60}Co , 25 to 55% of the total body burden is present in muscle tissue, with most of the remainder distributed among the liver, skin, and viscera.

Over 4000 fish were collected from the islands with a letter and number indicator shown in Figures 1 and 2. The majority of collection sites are in regions of the atolls with highest radionuclide concentrations in the sediment, and most of the fish were collected from these islands. Mean concentrations of the radionuclides are necessarily higher because more fishing occurred at these sites. Concentrations of the different radionuclides in flesh or muscle samples of the reef and pelagic fish are shown in Table 3. All concentrations measured in fish collected from the different islands of the atolls and the lagoon since 1976 were decay corrected to 10/98. The log normal mean concentration was then determined from the individual decay corrected values. This value represents the average

concentration of the specific radionuclide that might be expected in the flesh of any fish caught in 10/98 from the lagoons. In computing these values it was necessary to assume that the only loss of the radionuclides over the last 22 years was from radioactive decay only. A recent assessment (Noshkin et al., 1997c) of selected data shows that for some, but not for all radionuclides, the effective half life in the atoll environment is shorter than their radiological half life. Therefore, the values shown in Table 3 could be lower for some radionuclides. Mean concentrations are computed in the same manner for the flesh of mollusks and crustacean and are shown in Table 4.

Concentrations of ^{137}Cs in flesh and viscera of the fish are comparable but, because of the larger mass, most of the radionuclide accumulated by fish is found associated with the edible flesh; the lowest percentages are associated with bone and liver. Concentrations associated with surgeonfish were always greater than levels in flesh of goatfish, and generally exceed or were equivalent to the levels in mullet collected at the same time from different islands of the atolls. The surgeonfish are the better environmental indicators for ^{137}Cs levels. At Bikini, higher concentrations of ^{137}Cs were generally found in flesh of reef fish from the northwest quadrant of the atoll, and the lowest levels were associated with reef species from the eastern reef. At Enewetak, generally higher concentrations were measured in the reef fish from the northern half of the atoll and lowest levels were found associated with the fish from the southeastern and southern reef of the atoll. These patterns are consistent with the patterns of activity found associated with the sediments and demonstrate that these reef species are territorial. No significant difference in the mean concentration for ^{137}Cs is noted among the fish from either atoll, and reflects the similarity found in the mean concentrations in sediment and seawater at the two atolls. Based on the difference in mean sediment and seawater concentrations for $^{239+240}\text{Pu}$ found at Bikini and Enewetak, a similar large difference was expected in mean fish concentrations. It is difficult to conclude that there is any real difference in the mean muscle concentration in fish from Bikini compared to Enewetak, and supports earlier suggestions (Noshkin et al., 1981; 1987) that at these atolls the transuranics in some fish may be unrelated to the environmental concentrations.

Most striking were the differences found for ^{207}Bi concentrations in muscle tissue. Over 70% of the whole-body activity in goatfish and pelagic fish are associated with the muscle tissue, whereas less than 20% is found in the muscle of the mullet and surgeonfish. Mean concentrations in muscle for the 3 species of reef fish are summarized in Table 3. Concentrations associated with the flesh of goatfish are at least an order of magnitude greater than levels in surgeonfish or mullet collected at the same time from the same island. As with the difference found for ^{207}Bi concentrations in sediment, the mean concentration of ^{207}Bi in goatfish from Enewetak Atoll (excluding Enjebi

Island) slightly exceeds the value determined at Bikini. The highest levels were consistently found in muscle of goatfish collected on the reef of Enjebi Island (E-10), Enewetak Atoll, where the ^{207}Bi concentration in the muscle is more than a factor of 10 higher than the average over the rest of Enewetak Atoll and over Bikini Atoll. The highest ^{207}Bi concentrations are found in offshore sediments at Enjebi Island (see Fig. 2). Consequently, goatfish are clearly an excellent indicator species for ^{207}Bi levels in the lagoon environment.

Concentrations for the naturally occurring radionuclides, ^{210}Po and ^{210}Pb , in fish and invertebrates from the Marshall Islands (Noshkin et al., 1994) are shown in Tables 3 and 4. Dose from natural radioactivity will be compared to the dose from all man-made activity in the marine food ingestion pathway.

Dose Assessment

Exposure pathways at Bikini and Enewetak Atolls are ingestion of terrestrial foods, marine foods, and cistern/ground water; external gamma; and, inhalation of resuspended soil. Extensive data have been developed for each of these exposure pathways, and detailed dose assessments for returning populations have been provided for both atolls (Robison et al, 1987, 1997); details of the dose assessment methodology can be found in these references.

The marine pathway is of interest because most of the nuclear tests were conducted in or on the lagoon or reef thereby injecting a significant inventory of radionuclides into the sediment and water. Moreover, a major source of protein in the diet of Marshall Islanders comes from the consumption of fresh fish and other marine species obtained at the atolls. Consequently, the data for the radionuclide concentrations in fish, crustaceans, and invertebrates at Bikini and Enewetak have been used in conjunction with a diet model we use for the Marshall Islands (Robison et al., 1997), to estimate the dose to inhabitants at the atolls.

The total intake of marine foods in the diet model for the Marshall Islands is 0.56 kg per week, which accounts for about 5% of the locally derived food and liquid items, and about 3% of the total diet that includes imported foods. The marine foods include reef fish, pelagic fish, clams, turtles (and eggs), octopus, lobster, crabs, and marine feeding birds (and eggs). In Table 5 are listed the annual committed effective dose in 1998 for the radionuclides that contribute in any significant way to the dose at the atolls. Values are ranked from highest to lowest contributions at Enewetak. A different sequence applies at Bikini. The total dose for Enewetak Atoll is $0.0012 \text{ mSv y}^{-1}$ and that for Bikini Atoll $0.0030 \text{ mSv y}^{-1}$. At Bikini Atoll ^{137}Cs contributes about 33% of the total dose, while $^{239+240}\text{Pu}$ and ^{241}Am contribute about 43% and 12%, respectively. Most of the dose from $^{239+240}\text{Pu}$ and ^{241}Am is from the

consumption of Tridacna clams (and other types of clams) which concentrate the transuranic radionuclides to a much greater extent than fish and crustaceans. Bismuth-207 contributes about 3%.

The dose from ^{137}Cs at Enewetak Atoll accounts for about 54% of the total dose. Notice that the contribution from $^{239+240}\text{Pu}$ and ^{241}Am is much lower than at Bikini Atoll. This is because we have only two samples of Tridacana clams from the atoll that we know comes from an area of low concentration of $^{239+240}\text{Pu}$ in the water. Several Tridacana samples from around the lagoon are now available that will be analyzed in the very near future. The concentration of the transuranic radionuclides will undoubtedly be much higher as at Bikini Atoll thus increasing the contribution of $^{239+240}\text{Pu}$ and ^{241}Am to the total dose. This will nearly double the dose of $0.0012 \text{ mSv y}^{-1}$ listed in Table 5 to about $0.0023 \text{ mSv y}^{-1}$. Moreover, it will lower the percentage contribution of ^{137}Cs and increase the percentage contribution of the transuranic radionuclides. The results will be much more similar to those of Bikini Atoll listed in Table 5.

Enewetak Atoll is unique in that there is much more ^{207}Bi in the lagoon sediments, especially in the north end of the atoll near Enjebi Island. The contribution to the dose of ^{207}Bi is much higher (22.3%) at Enewetak Atoll than at Bikini Atoll (3%).

Inventories of $^{239+240}\text{Pu}$, ^{241}Am , ^{90}Sr , and ^{207}Bi are the largest of all the measurable radionuclides in the lagoon sediment. For comparison, the inventory of ^{137}Cs is much smaller, but leads to a dose of which is about 33% of the total estimated dose. About 90% of the total dose is delivered by ^{137}Cs , ^{90}Sr , $^{239+240}\text{Pu}$, ^{241}Am and ^{207}Bi . The dose for each radionuclide compared to its inventory in the lagoon sediments is summarized in Table 6. The dose per unit inventory for ^{137}Cs ranges from 0.14 to $0.16 \text{ } \mu\text{Sv h}^{-1} \text{ per GBq m}^{-2}$ at the two atolls. This is greater by about a factor of 20 to 50 than for $^{239+240}\text{Pu}$ and ^{241}Am . Even though the inventories of $^{239+240}\text{Pu}$ and ^{241}Am are quite large, they generate a far smaller dose than does a much smaller inventory of ^{137}Cs . The dose per unit inventory for ^{137}Cs is about 5 to 10 times that of ^{207}Bi .

We have also measured the concentration of naturally occurring ^{210}Po and ^{210}Pb in the marine species at the atolls (Noshkin et al., 1994). A comparison of the estimated dose from the natural radionuclides with the bomb-related radionuclides at Enewetak is shown in Table 5. The naturally occurring radionuclides dominate the effective dose from the consumption of fresh marine foods. The radionuclides generated from nuclear tests contribute between 0.1% and 0.3% of the total effective dose resulting from the marine pathways at the two atolls. However, as

mentioned previously, the dose at Enewetak will nearly double when the $^{239+240}\text{Pu}$ and ^{241}Am clam data are available, so the range will be 0.2 to 0.3%.

Over the next 15 to 100 years, the contribution from all the radionuclides other than $^{239+240}\text{Pu}$, ^{241}Am , and ^{238}Pu essentially will have disappeared. At that time, the dose from the consumption of marine foods or the remaining transuranium elements will be between 0.09% and 0.15% of the dose from $^{210}\text{Po} + ^{210}\text{Pb}$ (Table 7). $^{239+240}\text{Pu}$ contributes between 70–80% of the total, ^{241}Am about another 20–23%, and ^{238}Pu about 1–5%. Within about 350 y, only $^{239+240}\text{Pu}$ and ^{241}Am will be present. Consequently, the large inventories of $^{239+240}\text{Pu}$ and ^{241}Am in the atoll lagoons play a very minor role in the total dose received by people consuming marine fish, molluscs, crustaceans, and invertebrates at the atolls.

A detailed dose assessment that includes all exposure pathways has been made for Bikini Atoll (Robison et al., 1997) along with a detailed uncertainty analysis (Bogen et al., 1997). The relative contribution of each exposure pathway to the total effective dose is listed in Table 8. About 90% of the total effective dose results from ingestion of terrestrial foods, as a result of the uptake of ^{137}Cs from the soil into the edible fruit. Approximately another 10% of the dose is the result of external gamma exposure from ^{137}Cs in the soil. The rest of the pathways and radionuclides contribute about 1% of the total 50-y integral dose, and of that portion the contribution of the marine food chain is about 0.05%.

Several other gamma-emitting radionuclides such as $^{152, 154, 155}\text{Eu}$, ^{125}Sb , $^{101, 102}\text{Rh}$, ^{133}Ba , and $^{108\text{m}}\text{Ag}$ have been observed occasionally in other parts of fish and invertebrates such as in the viscera, bone and stomach contents, but never in the edible flesh. The total annual dose based on detection limits for these radionuclides in fish is approximately $2.3 \times 10^{-8} \text{ Sv y}^{-1}$, less than 3% of the total annual dose from the radionuclides shown in Table 5 from either atoll. Moreover, the radiological half-life of most of these radionuclides is short, and they will soon be nonexistent at the atolls, and the people do not eat the viscera, stomach contents, or bones.

Acknowledgments

Work performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

Reference

Adams CE, Farlow NH, Schell WR. The compositions, structures and origins of radioactive fall-out particles.

Geochim. et. Cosmochim. Acta 1960; 18: 42–56.

- Atkinson M., Smith SV., Stroup ED., Circulation in Enewetak Atoll lagoon. *Limnol. Oceanogr.* 1981; 26: 1074–1083.
- Bogen KT, Conrado CL, Robison WL. Uncertainty and variability in updated estimates of potential dose and risk at a U.S. nuclear test site—Bikini Atoll. *Health Phys.* 1997; 73: 115–126.
- Bonham K. Further contributions on gross beta radioactivity of biological and related samples at the Eniwetok Proving Ground, 1952-1958. University of Washington, Laboratory of Radiation Biology, Seattle, WA, UWFL-63, 1959.
- Emery KO, Tracy JJ, Ladd HS. Geology of Bikini and nearby atolls. Professional Paper 260-A, U.S. Geological Survey, U.S. Gov. Printing Office, Washington, DC, 1954; pp. 263.
- Gallery of U.S. Nuclear Tests. In: High energy weapons archive. (Carey Sublette administrator) (updated 12 Dec 1997), <http://www.envirolink.org/issues/nuketesting/hew/>.
- Hansen C. Nuclear weapons. The secret history. Aerofax, Inc. for Orion Books, Div. of Crown Pub. Inc., New York, New York, 1988.
- Hawthorne HA. Compilation of local fallout data from test detonations 1945–1962. Extracted from DASA 1251, Vol. II, Oceanic U.S. Tests, Report DNA 1251-2-EX, Defense Nuclear Agency, Washington, DC, 1979: pp. 333.
- Koranda JJ. Preliminary studies of the persistence of tritium and ^{14}C in the Pacific Proving Ground. *Health Phys.* 1965; 11: 1445–1457.
- Lowman FG. Marine biological investigations at the Eniwetok test site. In: Disposal of radioactive wastes, International Atomic Energy Agency, Vienna, Austria, 1960; 105–138.
- Lowman FG, Palumbo RF, South DJ. The occurrence and distribution of radioactive non-fission products in plants and animals of the Pacific Proving Ground. University of Washington, Applied Fisheries Laboratory, Seattle, WA, UWFL-51, 1957.
- McMurty GM, Schneider RC, Colin PL, Buddemeier RW, Suchanek TH. Redistribution of fallout radionuclides in Enewetak Atoll lagoon sediments by callianassid bioturbation. *Nature* 1985; 313: 674–677.
- Nelson V and Noshkin VE. Marine program. In: Enewetak survey report. U.S. Atomic Energy Commission, Nevada Operations Office, Las Vegas, NV; NVO-140, Vol. 1, 1973: 131–224.

- Nethaway DR, Barton GW. A compilation of fission product yields in use at the Lawrence Livermore Laboratory. Lawrence Livermore National Laboratory, Livermore, CA, Report UCRL-51458, 1973.
- Nevissi A, Schell WR. Distribution of plutonium and americium in Bikini Atoll lagoon. *Health Phys.* 1975; 28: 539–547.
- Noshkin V. Transuranium radionuclides in components of the benthic environment of Enewetak Atoll. In: Hanson WC, editor. *Transuranic elements in the environment*, DOE/TIC 22800, 1980, pp. 578–601.
- Noshkin VE, Wong KM, Eagle RJ, and Gatrousis C. Transuranics at Pacific atolls, IL. Concentration in the waters at Enewetak and Bikini. Lawrence Livermore National Laboratory, Livermore, CA, Report UCRL-51612, 1974.
- Noshkin VE, Eagle RJ, Wong KM, Jokela TA. Transuranic concentrations in reef and pelagic fish from the Marshall Islands. In: *Impacts of radionuclide releases into the marine environment*, International Atomic Energy Agency, Vienna, Austria, IAEA-AM-248/146, 1981: pp. 293–317.
- Noshkin VE, Wong KM, Eagle RJ, Robison WL. Comparative concentrations of ^{137}Cs , ^{90}Sr , $^{239+240}\text{Pu}$, and ^{241}Am in tissues of fish from the Marshall Islands and calculated dose commitments from their consumption. In: Pinder III JE, Alberts JJ, McLeod KW, Schreckhise RG, editors. *Environmental research on actinide elements*, CONF-841142, U.S. Dept. of Energy, Washington, D.C., 1987: 391–427.
- Noshkin VE, Robison WL, Wong KM. Concentration of ^{210}Po and ^{210}Pb in the diet at the Marshall Islands. *Sci. Total Environ.* 1994; 155: 87–104.
- Noshkin VE, Eagle RJ, Wong KM, Robison WL. Sediment studies at Bikini Atoll Part 2. Transuranium elements in surface sediments. Lawrence Livermore Laboratory, Livermore, CA, UCRL-LR-129379, 1997a.
- Noshkin, VE, Eagle RJ, Brunk JL, Robison WL. Sediment studies at Bikini Atoll Part 3. Inventories of some long-lived gamma emitting radionuclides associated with lagoon surface sediments. Lawrence Livermore Laboratory, Livermore, CA, UCRL-LR-129834, 1997b.
- Noshkin VE, Robison WL, Wong KM, Brunk JL, Eagle RJ, Jones HE. Past and present levels of some radionuclides in fish from Bikini and Enewetak Atolls. *Health Phys.* 1997c; 73(1): 49–65.
- Noshkin VE, Robison WL, Wong KM, Eagle RJ. Environmental behavior of plutonium isotopes in the marine environment of Enewetak Atoll. *J. Radioanal. & Nucl. Chem.* 1998a; 234(1–2): 243–249.

- Noshkin VE, Robison WL, Wong KM, Eagle RJ, Jokela TA, Brunk JL. Distribution and inventory of several long-lived radionuclides in Enewetak lagoon sediment. Lawrence Livermore Laboratory, Livermore, CA. 1998b (in preparation).
- Robison WL. Dose estimates for the marine food chain. In: Enewetak s Survey report. U.S. Atomic Energy Commission, Nevada Operations Office, Las Vegas, NV, NVO-140, Vol. 1, 1973, pp.526--541.
- Robison WL, Conrado CL, Phillips WA. Enjebi Island dose assessment. Lawrence Livermore National Laboratory, Livermore, CA, Report UCRL-53805, 1987.
- Robison WL, Bogen KT, Conrado CL. An updated dose assessment for resettlement options at Bikini Atoll—A U.S. nuclear test site. *Health Phys.* 1997; 73: 100–114.
- Schell WR, Lowman FG., Marshall RP. Geochemistry of transuranic elements at Bikini Atoll. In: Transuranic elements in the environment, Hansen W.C., editor. U.S. Dept. of Energy, Washington, DC, DOE/TIC-22800, 1980.
- Simon SL, Robison WL. A compilation of nuclear weapons test detonation data for U.S. Pacific Ocean tests. *Health Phys.* 1997; 73: 258–264.
- Von Arx WW. The circulation systems of Bikini and Rongelap lagoons. *Trans. Am. Geophys. Union* 1948;29: 861–870.
- Weiss HV, Shipman WH. Biological concentration by killer clams of cobalt-60 from radioactive fallout. *Science* 1957; 125: 695.
- Welander AD. Radioactivity in the reef fishes of Belle Island Eniwetok Atoll April 1954 to November 1955. University of Washington, Applied Fisheries Laboratory, WA. UWFL-49, 1957.
- Welander AD, Bonham K, Palumbo RF, Gessel SP, Lowman FG, Jackson WB, McClintock R, Lewis GB. Bikini-Eniwetok studies, 1964. Part II, Radiobiological Studies. College of Fisheries, University of Washington, Seattle, WA, Report UWFL-93, Pt. II, 1967.
- Welander AD. Distribution of radionuclides in the environment of Eniwetok and Bikini Atolls, August 1964. In: Nelson DJ, Evans FC, editors. *Proc. of 2nd National Symp. On Radioecology*, U.S. Atomic Energy Commission, Washington DC, CONF-670503, 1969, pp.346–354.
- Wong KM., Jokela TA, Eagle RJ, Brunk JL, Noshkin VE. Radionuclide concentrations, fluxes, and residence times at Santa Monica and San Pedro Basins. *Prog. Oceanog.* 1992; 30: 353–391.

Figure Captions.

Figure 1. Concentration contours (Bq kg^{-1}) for ^{241}Am associated with fine (<0.5 mm)[upper]and coarse (>0.5 mm) [lower] material in surface sediments in Bikini lagoon. Quantities in bold and underlined are total yields at the test site locations. The number of tests at the different locations is given in parenthesis. Island numbers identify reef fishing sites within the atoll.

Figure 2. Concentration contours (Bq kg^{-1}) for ^{241}Am [upper] and ^{207}Bi [lower] associated with fine (<0.5 mm) material in surface sediments in Enewetak lagoon. Quantities in bold and underlined are total yields at the test site locations. The number of tests at the different locations is given in parenthesis. Island numbers identify reef fishing sites within the atoll.

Table 1. Radionuclide inventories in Bikini and Enewetak lagoon sediment

Decay corrected to October 1998									
Radionuclide	Collection Date	TBq (doc) ^a to 2 cm	± error	TBq to 2 cm	GBq km ⁻² to 2 cm	TBq to 30 cm	± error	GBq km ⁻² to 30 cm	± error
Bikini Atoll (lagoon area 629 km ²)									
²⁴¹ Am ^b	Jan-73	7.7	1.0	10	16	79	32	126	51
	Jun-79	12	1.6	13	21	102	41	161	66
²⁰⁷ Bi	Jan-73	0.35	0.15	0.2	0.3	2	1	3	1
	Jun-79	0.30	0.050	0.2	0.3	2	1	2	1
¹³⁷ Cs	Jan-73	1.1	0.55	0.6	1	5	3	7	5
	Jun-79	0.85	0.30	0.5	1	4	2	6	3
⁶⁰ Co	Jan-73	1.6	0.45	0.05	0.1	0.4	0.2	0.6	0.3
	Jun-79	0.35	0.050	0.03	0.05	0.2	0.1	0.4	0.2
¹⁵⁵ Eu	Jan-73	11	1.5	0.3	0.5	2	1	4	1
	Jun-79	3.9	0.50	0.3	0.5	2	1	4	1
²³⁹⁺²⁴⁰ Pu	Jan-73	15	5.00	15	23	112	58	177	91
	Jun-79	17	2.30	17	27	132	53	209	85
Enewetak Atoll (lagoon area 933 km ²)									
²⁴¹ Am ^b	Jan-73	3.0	0.20	3	4	25	10	27	11
	Jun-79	2.9	0.20	3	3	24	9	26	10
²⁰⁷ Bi	Jan-73	3.8	0.30	2	2	17	7	18	7
	Jun-79	4.2	0.50	3	3	22	9	23	9
¹³⁷ Cs	Jan-73	1.2	0.30	0.7	1	5	2	6	3
	Jun-79	0.50	0.16	0.3	0.3	2	1	2	1
⁶⁰ Co	Jan-73	1.5	0.20	0.05	0.1	0.4	0.2	0.4	0.2
	Jun-79	0.36	0.16	0.03	0.03	0.2	0.1	0.2	0.1
¹⁵⁵ Eu	Jan-73	5.0	1.30	0.2	0.2	2	1	2	1
	Jun-79	1.4	0.20	0.1	0.1	0.8	0.3	0.8	0.3
²³⁹⁺²⁴⁰ Pu	Jan-73	8.1	0.30	8	9	62	24	67	26
⁹⁰ Sr	Jan-73	9.0	0.30	5	5	37	14	40	15

^a Doc is the date of collection.^b 1998 inventory is computed from decay and ingrowth from estimated levels of ²⁴¹Pu at the atolls (Noshkin, 1980).

Table 2. Summary of some concentrations for $^{239+240}\text{Pu}$ and ^{137}Cs in Seawater from within and outside Bikini and Enewetak Atolls

Location and Region Sampled Particulate	Month/Yr	# of Samples	Mean ²³⁹⁺²⁴⁰ Pu (Bq m ⁻³) ^a Solution Particulate	Mean ¹³⁷ Cs (Bq m ⁻³) ^a Solution
Enewetak Atoll				
Lagoon Water (Unfiltered)	Nov-72	37 ^b	1.16(01–2.6)	12.2(3.5–39)
Lagoon Water (Filtered)	Nov-72	estimate	0.69	12.2
Lagoon Water (Filtered)	Aug-74		33	1.23(0.2–2.6)
Lagoon Water (Filtered)	Feb-75			11(6–20)
Lagoon Water (Filtered)	May-76	19	0.55(0.1–1.3)	10(6–21)
Lagoon Water (Filtered)	Oct-79	7	0.75(0.3–1.3)	78% as oxidized species (+V,VI)
Lagoon Water (Filtered)	Jun-82	35	0.58(0.1–1.5)	78% as oxidized species (+V,VI)
Crater Water	Feb-75	7	8.4(1–40)	14(7–32)
Ocean reef water	May-76	9	0.90(0.2–2.6)	8(6–12)
Lagoon reef water	Nov-78	5	0.58(0.1–1.7)	15(9–23)
Lagoon reef water	Oct-79	4	1.37(0.1–2.3)	3.38(0.7–8.9)
Lagoon reef water	Feb-94	4	0.54(.13–70)	0.94(0.8–1.2)
Bikini Atoll				
Lagoon Water (Filtered)	Nov-72	16	1.42(0.1–2.9)	19(6–30)
Lagoon Water (Filtered)	Jan-77	26	1.83(0.7–3.9)	11(6–16)
Lagoon Water (Filtered)	Feb-81	9	1.24(0.4–3.4)	64 ± 15% as oxidized (+V,VI)
Lagoon Water (Filtered)	Sep-82	20	1.40(0.7–4.4)	90 ± 12% as oxidized (+V,VI)
Lagoon Water (Filtered)	Sep-82	20		12(5–20)
Lagoon reef water	Nov-78	8	1.04(0.4–1.8)	17(8–21)
Bravo Crater water	Sep-82	16	1.29(0.8–1.6)	96% as (+V,VI)
Outside Atolls				
North Equatorial				
Pacific Surface water	1972-1984	26 ^b	0.014 ± 0.004	5 ± 2
1-5 miles West & South of Bikini	Nov-72	5	0.55(0.1–1.3)	11(7–17)
1 mile South of Wide Pass, Enewetak	Apr-76	3	0.20(0.1–0.3)	5(4–7)

^a Range in concentrations is shown in parenthesis.

^b Unfiltered samples. All other samples are filtered through 0.45 or 1 micron filters.

Table 3. Log Normal Mean & Range in Fish Muscle Concentrations (Bq kg⁻¹ wet wt). All data decay corrected from collection date to 10/98 and then averaged (Number of samples are given in parenthesis).

Enewetak Atoll	Log normal mean	Range	Bikini Atoll	Log normal mean	Range
¹³⁷ Cs all reef fish	2.3(100)	.04–19.3	¹³⁷ Cs all reef fish	2.7(68)	.21–12
¹³⁷ Cs all pelagic fish	2.2(32)	0.63–9.4	¹³⁷ Cs all pelagic fish	5.2(12)	.25–9.6
⁶⁰ Co all reef fish	0.69(97)	.01–3.2	⁶⁰ Co all reef fish	0.77(68)	.032–3.3
⁶⁰ Co all pelagic fish	0.35(31)	.02–1.6	⁶⁰ Co all pelagic fish	0.46(12)	.014–1.2
²⁰⁷ Pb all reef fish	15(100)	.01–444	²⁰⁷ Pb all reef fish	2.1(66)	.01–32
²⁰⁷ Pb Goatfish (Enjebi Is.)	258(9)	3–444			
²⁰⁷ Pb Goatfish (rest of atoll)	12(13)		²⁰⁷ Pb Goatfish	7.0(18)	0.41–32
²⁰⁷ Pb Surgeonfish	0.51(34)	.01–1.9	²⁰⁷ Pb Surgeonfish	0.27(12)	.014–.85
²⁰⁷ Pb Mullet	0.35(35)	.02–1.8	²⁰⁷ Pb Mullet	0.24(32)	.010–1.6
²⁰⁷ Pb Other reef fish	1.1(9)	.04–8.8	²⁰⁷ Pb Other reef fish	0.67(4)	.02–.85
²⁰⁷ Pb all pelagic fish	6.9(32)	.06–28	²⁰⁷ Pb all pelagic fish	3.2(12)	.17–7.7
²³⁹⁺²⁴⁰ Pu all reef fish	0.012(68)	.0004–.17	²³⁹⁺²⁴⁰ Pu all reef fish	0.013(39)	.0002–.041
²⁴¹ Pu ^b all reef fish	0.006		²⁴¹ Pu ^b all reef fish	0.036	
²³⁸ Pu all reef fish	0.0019(52)	.00005–.019	²³⁸ Pu all reef fish	0.0006(32)	.00002–.004
²³⁹⁺²⁴⁰ Pu all pelagic fish	0.0021(16)	.00003–.009	²³⁹⁺²⁴⁰ Pu all pelagic fish	0.0023(6)	.0005–.002
²⁴¹ Pu ^b all pelagic fish	0.001		²⁴¹ Pu ^b all pelagic fish	0.0065	
²³⁸ Pu all pelagic fish	0.00094(6)	.00007–.002	²³⁸ Pu all pelagic fish	0.0003(1)	
²⁴¹ Am all reef fish	0.0050(11)	.0008–.014	²⁴¹ Am all reef fish	0.0072(13)	.0004–.039
²⁴¹ Am all pelagic fish	0.0024(6)	.0008–.008	²⁴¹ Am all pelagic fish	0.0018(2)	.001–.002
⁹⁰ Sr all reef fish	0.02(1)		⁹⁰ Sr all reef fish	0.045(8)	.0096–.13
⁹⁰ Sr all pelagic fish	no data		⁹⁰ Sr all pelagic fish	0.0053(1)	
²⁴⁴ Cm all reef fish	0.00029(2)	.00005–.0005	²⁴⁴ Cm all reef fish	no data	
^{113m} Cd all reef fish	0.093(4)	.033–.20	^{113m} Cd all reef fish	<.06	
^{113m} Cd all pelagic fish	<.06		^{113m} Cd all pelagic fish	<.06	
⁹⁹ Tc all reef fish	no data			0.0009(1)	
²¹⁰ Po all reef fish ^a	12.5 (73)		²¹⁰ Pb all reef fish ^a	1.0(27)	
²¹⁰ Po all pelagic fish ^a	16.4(19)		²¹⁰ Pb all pelagic fish ^a	2.6	

^a Data from Noshkin et al., 1994.

^b No direct measurement in fish. Values estimated from mean ²⁴¹Pu/²³⁹⁺²⁴⁰Pu activity (decay corrected) ratios in the environments of Bikini and Enewetak (Noshkin, 1980) and multiplied by the mean ²³⁹⁺²⁴⁰Pu concentration in fish.

Table 4. Radionuclide concentration data for flesh of invertebrates collected from Enewetak and Bikini Atolls between 1972 and 1991.

Ln Normal Mean Flesh Concentrations (Bq kg ⁻¹ wet wt) for Molluscs and Crustaceans. All data decay corrected from collection date to 10/98 and then averaged. (Number of samples is given in parenthesis.)		
Radionuclide	Molluscs	Crustacean
Enewetak		
²³⁹⁺²⁴⁰ Pu	.032 ± .024(4) ^c	.0038 ± .0041(3)
²⁴¹ Pu ^b	0.018	0.002
²³⁸ Pu	.0023 ± .0022(3)	.00016 ± .00015(3)
¹³⁷ Cs	<.09 ± .08(4)	.069 ± .037(3)
⁶⁰ Co	1.84 ± 1.89(4)	.122 ± .024(3)
²⁰⁷ Bi	0.15 ± .05(2)	.048 ± .012(3)
²⁴¹ Am	.010 ± .005(4)	<0.1(3)
⁹⁰ Sr	.025 ± .019(4)	no data
Bikini		
²³⁹⁺²⁴⁰ Pu	0.92 ± .76(9)	.072 ± .073(5)
²⁴¹ Pu ^b	2.61	0.20
²³⁸ Pu	.019 ± .021(7)	<.002(5)
¹³⁷ Cs	<.36 ± .43(12)	.44 ± .54(3)
⁶⁰ Co	4.9 ± 6.0(12)	.34 ± .02(2)
²⁰⁷ Bi	0.26 ± .29(10)	<0.2(3)
²⁴¹ Am	.32 ± .29(7)	<0.1(3)
⁹⁰ Sr	<.04 ± .02(2)	no data
Both Atolls		
²¹⁰ Po ^a	56(6)	10.1(2)
²¹⁰ Pb ^a	1.6(2)	0.15(2)

^a Data from Noshkin et al., 1994.

^b No direct measurement in invertebrates. Values estimated from mean ²⁴¹Pu/²³⁹⁺²⁴⁰Pu activity (decay corrected) ratios in the environments of Bikini and Enewetak (Noshkin, 1980) and multiplied by the mean ²³⁹⁺²⁴⁰Pu concentration in mollusc and crustacean.

^c Four samples of muscle and mantle tissues were separated from 2 clams.

Table 5. The annual committed effective dose at Enewetak Atoll in 1998 from the consumption of marine foods.

Radionuclide ($T_{1/2}$)	Committed effective dose mSv y^{-1}	
	Enewetak	Bikini
Bomb-related		
^{137}Cs (30.1 y)	6.5×10^{-4} (54%)	9.8×10^{-4} (33%)
^{207}Bi (38 y)	2.7×10^{-4} (22.5%)	9.3×10^{-5} (3.1%)
$^{239+240}\text{Pu}$ (24065 y, 6537 y)	9.2×10^{-5} (7.7%) ^a	1.3×10^{-3} (43%)
^{60}Co (5.27 y)	7.6×10^{-5} (6.3%)	1.4×10^{-4} (4.7%)
$^{113\text{m}}\text{Cd}$ (13.6 y)	5.2×10^{-5} (4.3%)	4×10^{-5} (1.3%)
^{241}Am (432.2 y)	3.0×10^{-5} (2.5%)	3.7×10^{-4} (12%)
^{90}Sr (29.12 y)	2.5×10^{-5} (2.0%)	1.7×10^{-5} (0.57%)
^{238}Pu (87.74 y)	1.0×10^{-5} (0.80%)	1.9×10^{-5} (0.63%)
^{241}Pu (14.4 y)	8.8×10^{-7} (0.07%)	5.5×10^{-5} (1.8%)
	Total	0.0012 (100%)
		0.0030 (100%)
Naturally occurring		
^{210}Po (138.38 d)	0.96	0.96
^{210}Pb (22.3 y)	0.09	0.09
	Total	1.1
		1.1

^a Mollusc data for Enewetak consists of only 2 samples from one location. More data will be available in the near future, and the transuranium radionuclide contribution to dose will be revised upward. Data from Bikini Atoll indicates that the mean concentration of transuranium radionuclides is higher in molluscs than in reef fish, and consequently, the transuranium radionuclides contribute a larger percentage of the total dose in the marine pathway that rivals the ^{137}Cs contribution.

Table 6. The annual effective dose from marine foods per radionuclide inventory to 30 cm depth in lagoon sediments.

Radionuclide	Dose per unit inventory $\mu\text{Sv y}^{-1}/\text{GBq m}^{-2}$	
	Enewetak	Bikini
^{137}Cs	0.16	0.14
$^{239+240}\text{Pu}$	0.0014	0.0067
^{241}Am	0.0011	0.0026
^{207}Bi	0.013	0.031
^{90}Sr	0.00063	—

Table 7. The annual committed effective dose for bomb-related transuranium radionuclides as a percentage of the dose from naturally occurring ^{210}Po and ^{210}Pb from consumption of marine foods.

Radionuclide	Percent of $^{210}\text{Po}+^{210}\text{Pb}$ annual committed effective dose	
	Enewetak	Bikini
$^{239+240}\text{Pu}$	0.008	0.12
^{241}Am	0.003	0.034
^{238}Pu	0.0007	0.000002
Total	0.012 ^a	0.15

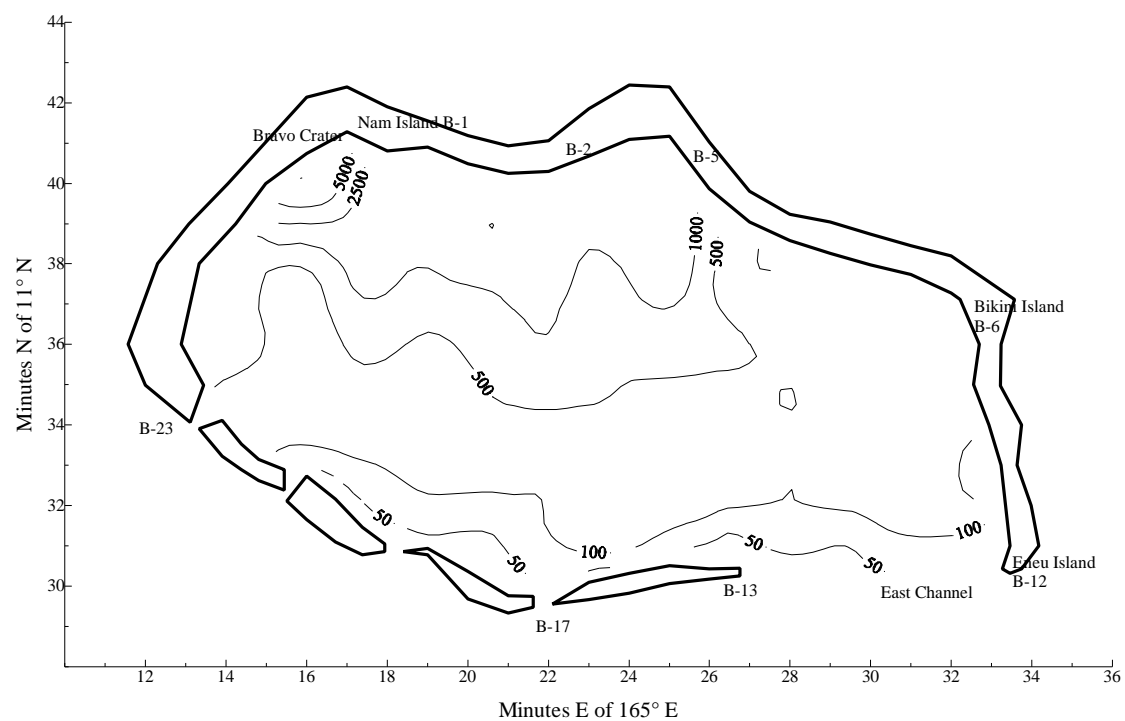
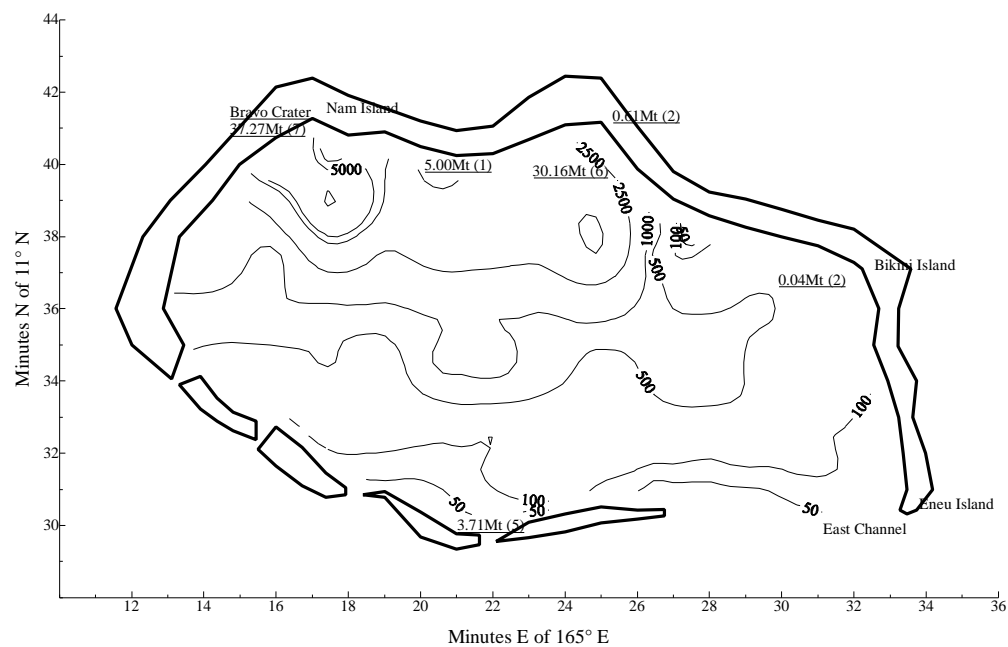
^a This number is expected to be in the range of 0.09 to 0.10 when the clam data are available.

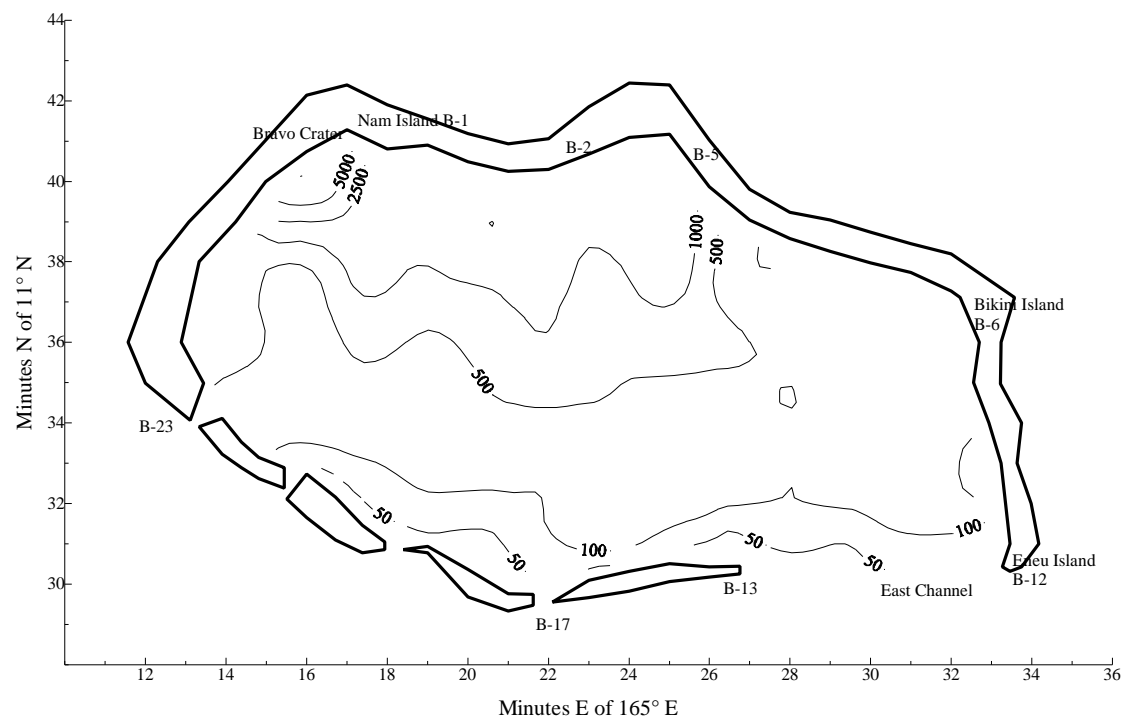
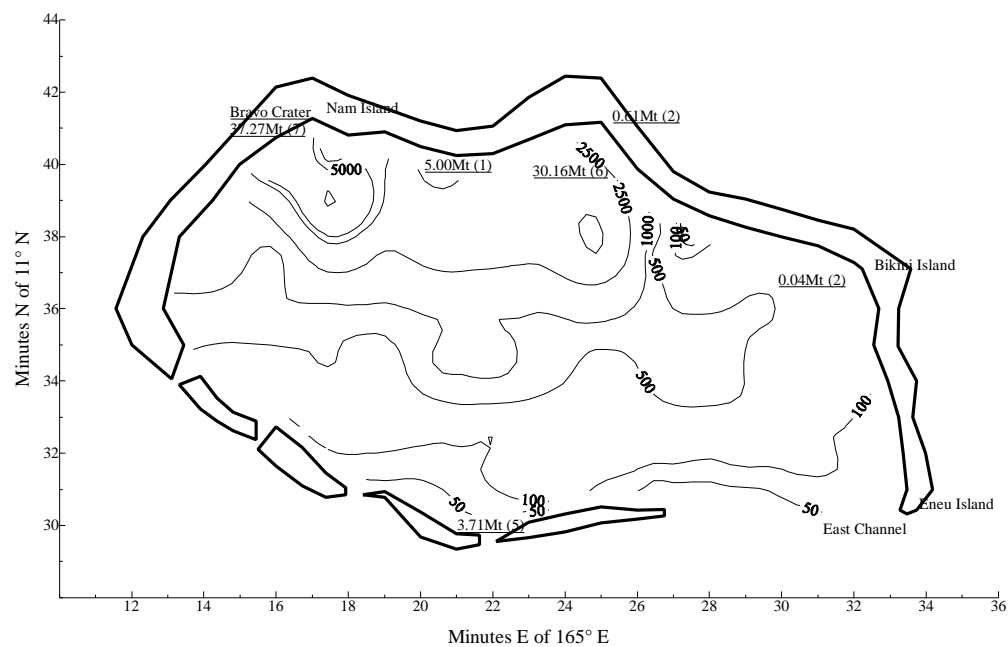
Table 8. The 30-, 50-, and 70-y integral effective dose for the various exposure pathways for the imported foods available diet^a at Bikini Atoll.

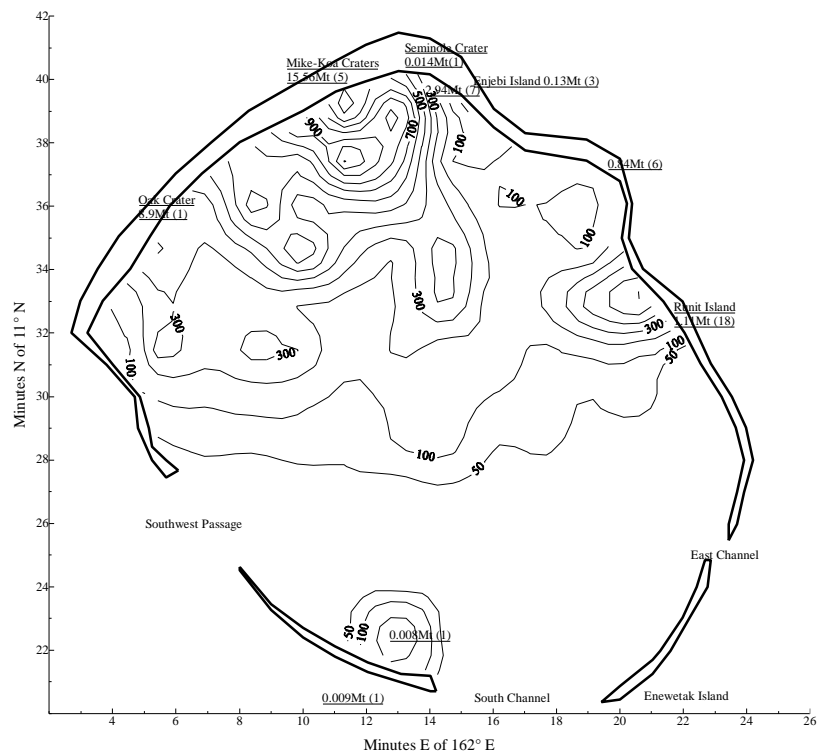
Exposure Pathway	Effective integral equivalent dose, mSv		
	30 y	50 y	70 y
Terrestrial food	82	110	130
External gamma	9.1	13	15
Marine food	0.048	0.096	0.16
Cistern and ground water	0.15	0.21	0.25
Inhalation	0.12	0.27	0.38
Total ^b	91	130	150

^a The imported foods available diet is based on a total intake that is comprised of 60% imported foods and 40% locally grown crops.

^b The total dose may vary in the second decimal place due to rounding.







01

